

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
BRUCE H BERSTED, ET AL. : EXAMINER: WOODWARD, A.L.
SERIAL NO: 10/531,251 :
FILED: OCTOBER 7, 2005 : GROUP ART UNIT: 1796
FOR: ANTI-YELLOWING :
POLYCONDENSATION POLYMER
COMPOSITIONS AND ARTICLES

DECLARATION

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

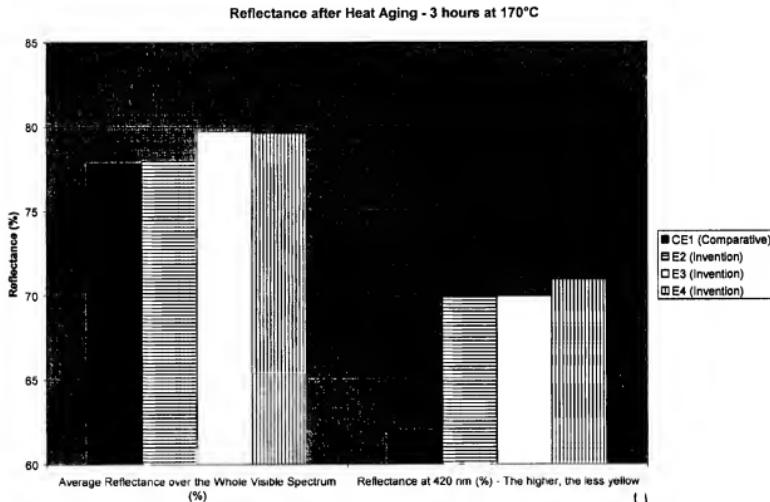
SIR:

Now comes Henri Massillon, who deposes and states as follows:

1. That I am an inventor of the above-identified application for patent in the United States, and that I understand English.
2. That it is my understanding that the composition claims pending in this application require the presence of a white pigment in a concentration of at least 4 weight %, based on the total weight of the composition, and a black pigment present in a concentration of at least 0.0001 weight % and up to 0.002 weight %, (i.e., 20 ppm) based on the total weight of the composition.
3. That in Example 1 beginning at specification page 18, polyphthalamide, a partially aromatic polyamide, was blended with white pigment and 0, 9, 12 or 16 ppm carbon black, and heat tested for yellowing. The results, presented in Table form at pages 21-22 of the



original specification are graphed below and show a substantial, unexpected, and surprising improvement with respect to yellowing: while the average reflectance over the whole visible spectrum remains constant or shows improvement upon the addition of the presently claimed limited amounts of black pigment, the average reflectance at those wavelength characteristic of yellowing show dramatic improvement when the invention combination of up to 20 ppm black pigment is used in combination with white pigment:



The graph above uses the data from Tables 4 and 5 of the specification and plots the reflectivity of CE1, the comparative example containing no black pigment, and of E2-E4, Examples according to the invention containing 9-16 ppm black pigment. Two groups of data are shown: average reflectance over the whole visible spectrum and average reflectance at 420 nm, characteristic of yellowing. As is clear from the data in the original specification, graphed above, the present invention composition using a very small but specific amount of black pigment (i.e., up to 20 ppm) provides substantial, unexpected, and surprising anti-yellowing properties.

4. In addition, a new composition (E1) has been prepared in accordance with the invention containing polyarylethersulfone, a white pigment, and a black pigment, namely "MPC channel black" carbon black present at 0.001 wt. % based on the total weight of the composition (i.e., 10 ppm). This composition was compared to a composition identical thereto but for the lack of the carbon black (C1).

Specifically, composition (E1) in accordance with the invention was prepared consisting of:

- *a commercial polyarylethersulfone, namely Udel® P-1700 NT11 commercially available from SOLVAY ADVANCED POLYMERS, L.L.C. having a heat deflection temperature of about 174°C under a load of 1.82 MPa when measured according to ASTM D648 ;
- *a white pigment, namely Kronos® 2230 TiO₂ – 40 wt. % based on the total weight of the composition ;
- *a black pigment, namely "MPC channel black" carbon black – 0.001 wt. % based on the total weight of the composition.

For comparison purposes, composition (CE1) free of black pigment was prepared consisting of :

- *a commercial polyarylethersulfone, namely Udel® P-1700 NT11 commercially available from SOLVAY ADVANCED POLYMERS, L.L.C. having a heat deflection temperature of about 174°C under a load of 1.82 MPa when measured according to ASTM D648 – same lot as the lot used for example E1 ;

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▪ a white pigment, namely Kronos® 2230 TiO₂ – 40 wt. % based on the total weight of the composition – same lot as the lot used for example E1.

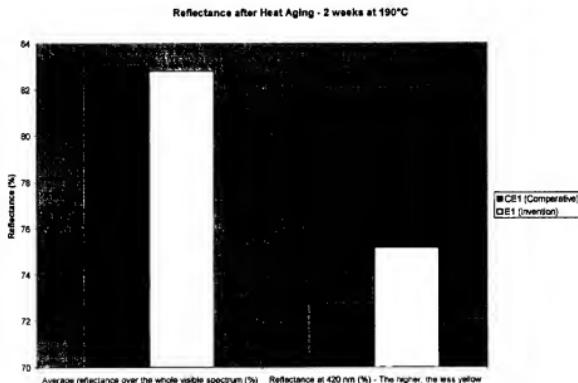
E1 and C1 were submitted to identical heat aging conditions, as follows : 190°C, room humidity.

After heat aging the reflectance of compositions E1 and C1 were measured at different times (non aged, aged after one week at 190°C, and aged after 2 weeks at 190°C) under the same conditions and using the same techniques as those described in the above-noted patent application. The results are presented in Table I below.

Table I : Measured reflectances

Wavelength, nm	E1 non aged	CE1 non aged	E1 Aged after 1 week at 190°C	CE1 Aged after 1 week at 190°C	E1 Aged after 2 weeks at 190°C	CE1 Aged after 2 weeks at 190°C
360	11.242	11.844	11.502	11.49	11.54	11.428
370	12.496	13.094	12.664	12.636	12.686	12.547
380	15.907	16.558	15.937	15.883	15.878	15.705
390	25.074	25.819	24.798	24.742	24.79	24.263
400	44.857	45.678	43.857	43.689	42.986	42.257
410	69.369	70.553	66.63	66.178	64.446	62.719
420	83.12	84.756	78.436	78.758	72.199	72.701
430	86.607	88.75	81.462	80.836	78.073	75.457
440	87.175	89.92	82.305	81.831	79.039	76.523
450	87.345	89.713	82.577	82.522	79.674	77.28
460	87.441	89.968	83.226	83.085	80.204	77.968
470	87.494	90.11	83.594	83.633	80.778	78.691
480	87.599	90.332	84.017	84.256	81.378	79.517
490	87.656	90.471	84.415	84.879	82.042	80.439
500	87.704	90.663	84.872	85.589	82.754	81.437
510	87.766	90.788	85.236	86.202	83.427	82.41
520	87.834	90.949	85.588	86.856	84.038	83.345
530	87.907	91.115	85.965	87.475	84.699	84.277
540	87.995	91.299	86.322	88.111	85.285	85.213
550	88.041	91.434	86.66	88.704	85.819	86.049
560	88.052	91.522	86.881	89.171	86.265	86.775
570	88.112	91.683	87.105	89.625	86.635	87.433
580	88.202	91.834	87.333	90.077	87.015	88.07
590	88.226	91.942	87.508	90.433	87.304	88.594
600	88.242	92.059	87.636	90.77	87.578	89.129
610	88.231	92.124	87.7	91.006	87.707	89.476
620	88.279	92.243	87.825	91.277	87.923	89.908
630	88.297	92.336	87.901	91.497	88.066	90.241
640	88.314	92.425	87.992	91.767	88.215	90.611
650	88.341	92.515	88.034	91.94	88.333	90.888
660	88.352	92.581	88.089	92.115	88.415	91.124
670	88.319	92.604	88.074	92.194	88.409	91.271
680	88.3	92.674	88.053	92.284	88.429	91.417
690	88.288	92.705	88.064	92.383	88.446	91.538
700	88.231	92.715	88.041	92.483	88.467	91.711
710	88.178	92.704	87.976	92.497	88.441	91.817
720	88.162	92.758	88.015	92.631	88.458	91.949
730	88.105	92.753	87.979	92.653	88.385	92.029
740	87.875	92.555	87.756	92.505	88.215	91.93
750	87.617	92.38	87.518	92.343	87.964	91.813

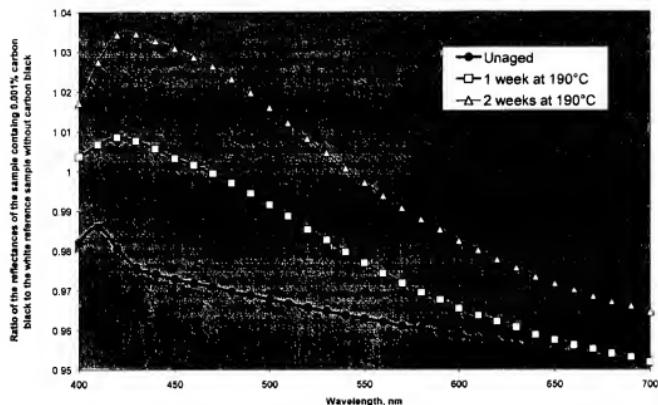
These results again show a substantial, unexpected, and surprising improvement with respect to yellowing: while the average reflectance over the whole visible spectrum remains essentially constant upon the addition of the presently claimed limited amount of black pigment, the average reflectance at those wavelength characteristic of yellowing show dramatic improvement when 10 ppm black pigment is used in combination with white pigment, as claimed:



In addition, when the data in Table I above is graphed, as in the Figure below, using the ratio of the measured reflectance at different wavelengths covering the visible spectrum (400-700nm) the results obtained at low wavelengths of the visible spectrum, in particular those obtained in the neighborhood of 420 nm corresponding to yellow light, show unexpectedly superior results achieved with composition EI after heat aging:

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Reflectance ratio E1/CE1



This ratio graphed above, E1/CE1, is $(\text{Reflectance}_{\lambda, \text{E1}} / \text{Reflectance}_{\lambda, \text{CE1}})$, wherein λ is the wavelength. A ratio greater than 1 at a given wavelength indicates that the composition according to the invention comprising the black pigment (E1) exhibits a reflectance greater than the reflectance of the comparative composition CE1 free of black pigment at this wavelength. Low wavelengths (400-550 nm) are particularly important; in particular wavelengths in the neighborhood of 420 nm correspond to yellow light: in the above graph the higher the reflectance ratio, the less yellow the composition of the invention is as compared to CE1.

5. The examples in the specification as filed, using a partially aromatic polyamide, and the above additional experiments, using a polyarylethersulfone, use a range of polycondensation polymers that are so diverse and unrelated that they show that the unexpected and surprising results obtained for these polymers would be provided by all the

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polycondensation polymers selected from at least partially aromatic polyamides, polyamideimides, liquid crystalline polymers, polyimides, polyetherimides, polyaryletherketones, polyarylethersulfones, and polyphenylene sulfides. For example, the newly-presented results above were obtained for an amorphous polymer, while a semi-crystalline polymer was used in the set of examples contained in the original patent application, each polycondensation polymer tested having substantially different and unique heat deflection temperatures (about 120°C for the partially aromatic polyamide, to be compared to about 174°C for the polyarylethersulfone), these basis polymers also differing substantially in polarity, chemical resistance, and other physical properties. In this regard, the totality of the data provided demonstrates that the surprising and unexpected technical effects shown were not obtained by chance, or limited to only the tested species, and that this surprising and unexpected technical effect can be observed on highly different, almost opposite, engineering polymers.

6. That I am familiar with Ohtomo, U.S. 5,760,125, and the compositions described therein which are very limited with regard to morphology, requiring that the PPE phase be dispersed as a particulate in the PA phase, and that the pigment be dispersed in the PPE phase. Ohtomo requires both a polyphenylene ether resin and a polyamide resin dispersed in a particular morphology relative to one another, and the presence of certain components, including the pigment, only in the polyphenylene ether phase.

7. Ohtomo discloses the use of substantially more carbon black as compared with the presently claimed limited amount of 20 ppm. See Table I of the reference at column 11 where pigment (c) represents carbon black (note column 9, lines 59-60 of the reference). Here, the lowest amount of carbon black used is 0.05 wt.%, which translates to 500 ppm carbon black.

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8. The use in Ohtomo of substantially more carbon black as compared with that amount presently claimed is indicative of certain substantially more highly colored compositions than those of the present invention, limited to a maximum of only 20 ppm black pigment. There is no motivation in Ohtomo to use such little amount of black pigment - to the contrary, one skilled in the art would have understood that reducing so drastically the amount of carbon black would imply switching from an intense gray coloration as provided to an essentially pure white coloration, which by essence is much more sensitive to yellowing when submitted to heat and/or light aging. Ohtomo's exemplified compositions probably contain such a high amount of carbon black (from 500 to 2000 ppm) to *mask* the problem the present invention solves: the yellowing that results from the aging of the polymer. In Ohtomo, by providing at least gray-colored products, this problem is avoided. The reference thus does not suggest reducing the amount of carbon black as doing so would highlight the yellowing problem. Certainly nothing in the reference suggests that using only 20 ppm or less of black pigment could or would *solve* the problem, as has been demonstrated above.

9. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

November 13, 2009
DATE


Henri Massillon